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journal or publication title	The science reports of the Tohoku University. Ser. 8, Physics and astronomy
volume	25
number	1
page range	191-192
year	2004-12-14
URL	http://hdl.handle.net/10097/26206

Optical properties of excitons in ZnO-based quantum wells

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ZnO is a wide band gap (about 3.37 eV at room temperature) semiconductor. The experimental observation of room temperature lasing of ZnO thin films grown on sapphire substrate by laser-ablation molecular beam epitaxy method in recent year, started to attract considerable research attention because of the exploration of its potential application in ultra-violet light emitting diodes or laser diodes. Binding energy of exciton in ZnO is about 60 meV, so that possesses the ability to be stabilized at higher temperature. Therefore, for ZnO, expectation of room temperature exciton-related stimulated emission, which should have low pumping threshold than that of electron-hole plasma, is reasonable. Recently, sufficiently high-quality of ZnO quantum wells have been able to be obtained by employment of lattice-matched substrates (ScAlMgO4) instead of sapphire substrates. The stimulated emission induced by inelastic exciton-exciton scattering occurred throughout the temperature range from 5 K to room-temperature is demonstrated in ZnO/ZnMgO quantum well structures. However, it is believed that in principle the threshold for laser action using biexcitons as its mechanism is even lower that that using the inelastic exciton-exciton scattering process. Therefore, from the viewpoint of the application as a low-threshold semiconductor laser, a detailed study of biexciton-related processes is necessary for ZnO-based quantum well. Therefore, in this research, a detailed study of the linear as well as nonlinear optical properties of excitons in these two-dimensional systems was carried out in order to understand the optical phenomenon of these heterostructures, and also in view of their application to ultra-violet blue optoelectronic devices.

Two-set of ZnO-based quantum well samples investigated in this research were fabricated in Kawasaki laboratory (IMR. Tohoku Univ.) by Dr. A. Ohtomo, using combinatorial laser molecular beam laser system. By switching the mask patterns and targets during pulsed laser deposition, nine different films having various well-width of 0.7, 0.9, 1.3, 1.8, 2.4, 2.8, 3.7, 4.2, 4.7 nm, were grown on $ScAlMgO_4$ substrate for 10-periods. 5nm- thick $Zn_{1-x}Mg_xO$ alloy with x=0.12 and 0.27 were used as barriers here.

In order to understand the fundamental optical transition of these quantum well structures, absorption and photoluminescence measurement are employed to two-sets of $ZnO/Zn_{1-x}Mg_xO$ (x=0.12, 0.27) quantum wells. The Mg content of the barrier layer for x=0.12, 0.27 corresponds to barrier height of about 0.2, 0.5 eV respectively. Band edge absorption spectra exhibit strong excitonic features related to the transition of ground state exciton corresponding to n=1 subband. Quantum-confinement effects due to well-width of ZnO layer and barrier height of ZnMgO with different Mg contents are identified. Stokes shifts of the emission bands in ZnO quantum wells are well-width dependent. Therefore, in order to investigate the recombination dynamics of excitons, time-resolved photoluminescence measurement were carried out. The spectral distribution of decay time constants shows that emissions of these ZnO quantum wells are due to the radiative recombination of excitons localized at potential fluctuation.

The anomalies in absorption and photoluminescence spectra of ZnO/ZnMgO quantum wells with higher Mg contents and well-width with 4.2 nm and 4.7 nm are investigated by time-resolved photoluminescence measurement and excitation-density dependence of emission bands. We concluded that built-in internal electric fields, which induced by piezoelectric and spontaneous polarization, exist in these two quantum wells.

Localized biexciton emission has been reported in $ZnO/Zn_{0.73}Mg_{0.27}O$ quantum well. However, because of a strong localization effect existed in the ZnO-quantum well, the biexciton binding energy is unable to determine precisely from the photoluminescence studies. Therefore, a nanosecond nondegenerate pump-probe measurement were performed to four $ZnO/Zn_{0.73}Mg_{0.27}O$ quantum well samples with well-width of 3.7, 2.8, 2.4, 1.8 nm at 77 K. Lineshape of the differential absorption spectra is simulated by nonlinear absorption spectra considering the saturation of excitonic states. Biexciton formation by induced absorption from free exciton states to free biexciton states was identified. The well-width dependence of biexciton binding energy in ZnO/ZnMgO quantum well structure was determined. It should be pointed out that the quantum size effect on the biexciton binding energy of ZnO quantum well structure is demonstrated. For ZnO quantum well with well-width of 1.8 nm, the biexciton binding energy is about 26 meV. This implies that at room-temperature biexciton in ZnO quantum well is possible to be existed. Therefore, from the application view of points, biexciton-related lasing action, which practically should have lower threshold than inelastic exciton-exciton scattering, electron-hole plasma, that reported so far, may have the chance to be achieved in ZnO quantum well structures.